

VI.2 Lanthanum Gallate Electrolyte Based Intermediate-Temperature Solid Oxide Fuel Cell Development

S. (Elango) Elangovan

Ceramatec, Inc.

2425 South 900 West

Salt Lake City, UT 84119-1517

Phone: (801) 978-2162; Fax: (801) 972-1925; E-mail: Elango@ceramatec.com

DOE Project Manager: Lane Wilson

Phone: (304) 285-1336; E-mail: Lane.Wilson@netl.doe.gov

Subcontractors:

Sandia National Laboratory, Albuquerque, New Mexico, contact: Dr. Ron Loehman

New Mexico Tech, Socorro, New Mexico, contact: Prof. Deidre Hirschfeld

Objectives

- Evaluate alternative anode materials in order to reduce anode-electrolyte reactivity
- Develop tape cast process to fabricate thin electrolyte cells
- Fabricate single cells using a supported structure
- Demonstrate intermediate-temperature fuel cell operation
- Test short stacks using 10x10 cm cells
- Evaluate mechanical properties of gallate material

Approach

- Modify the anode composition and verify reduction in reactivity using x-ray diffraction of reacted anode-electrolyte powder mixture
- Perform tape sintering studies to fabricate supported single cells
- Test single cells at 700 – 800°C for short-term and long-term performance
- Measure strength of gallate bars

Accomplishments

- Determined that the modification introduced into the nickel-based anode reduced the reactivity between nickel and lanthanum gallate
- Fabricated single cells using thin supported electrolyte with electrolyte thickness ranging from 30 to 75 microns
- Demonstrated single-cell performance with an area-specific resistance of 0.5 ohm-cm² at 700°C
- Demonstrated stable 2,500-hour performance at an operating temperature of 700°C
- Characterized mechanical strength of gallate at room temperature and at 800°C

Future Directions

- Fabricate full-size (10x10 cm) thin electrolyte cells
- Perform stack tests to verify performance and stability benefits demonstrated in single cells
- Evaluate alternative cathode materials for intermediate-temperature operation

Introduction

Reducing the operating temperature of solid oxide fuel cells (SOFCs) offers several benefits: improvement in long-term stability by slowing physical and chemical changes in the cell materials; lower-cost systems by the use of less expensive balance of plant components; compatibility with hydrocarbon reformation process, allowing partial internal reformation, which in turn reduces the heat exchanger duty; and, finally, the potential to improve thermal cycle capability. In addition, the use of stainless steel interconnects is facilitated by the lower operating temperature. A temperature range of 650 to 700°C is ideally suited to derive the performance stability, system integration and cost benefits.

In order to derive the advantages of the lower operating temperature, two factors that limit the cell performance, namely the electrolyte resistance and electrode polarization, must be addressed. Lanthanum gallate compositions have shown high oxygen ion conductivity when doped with Sr and Mg. Unlike other oxygen ion conductors such as ceria and bismuth oxide that are potential candidates for lowering cell operating temperature, the Sr- and Mg-doped lanthanum gallate (LSGM) compositions are stable over the oxygen partial pressure range of interest. The combination of stability in fuel gas environment and the high oxygen ion conductivity makes the LSGM material a potential choice for intermediate-temperature SOFCs. However, challenges in the development of electrode materials and cell fabrication processes need to be overcome to make use of the potential of the LSGM electrolyte.

Approach

The nickel-based anode has been successfully demonstrated to be catalytic for fuel oxidation in zirconia electrolyte based SOFC systems. A modified nickel anode composition was evaluated using powder mixtures of the anode and LSGM electrolyte. The reacted powder mixture was analyzed using x-ray diffraction technique. Additionally, an 8-cell stack was tested for over 1,000 hours, and the anode-electrolyte interface was analyzed using scanning electron microscopy for evidence of nickel diffusion into the electrolyte.

Tape cast process development was performed to cast LSGM tape of various thicknesses to provide sintered electrolyte thicknesses ranging from 75 to 300 microns. The process variables included powder surface area, organic content in the tape slip, and sintering temperature. The primary objectives of the activity were to achieve sintered electrolyte density and flatness required for stacking. Single cells with 1 to 2.5 cm² active area were tested for performance characteristics and long-term stability.

Mechanical property evaluation was done at the Sandia National Laboratories to measure electrolyte strength after subjecting bar samples to various exposure and thermal cycle operations.

Results

The modification to the nickel anode was found to significantly reduce the reactivity with the LSGM electrolyte. X-ray diffraction pattern for the powder mixture of the anode-electrolyte calcined at 1,300°C is shown in Figure 1. Diffraction pattern for the baseline LSGM, NiO, and a mixture of the two are also shown in the figure. It can be seen that the reaction phase peaks prominent in the baseline mixture are significantly reduced in their intensities for the modified anode.

In order to verify the long-term stability of the anode-electrolyte interface, a stack was built and tested for 1,200 hours. Although performance degradation was observed, attributable to chromium evaporation from the interconnect, scanning electron

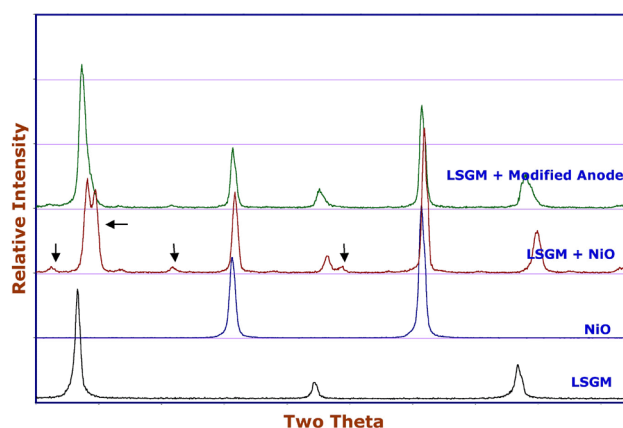


Figure 1. Powder X-ray of Baseline and Reacted Electrolyte, Anode Powders

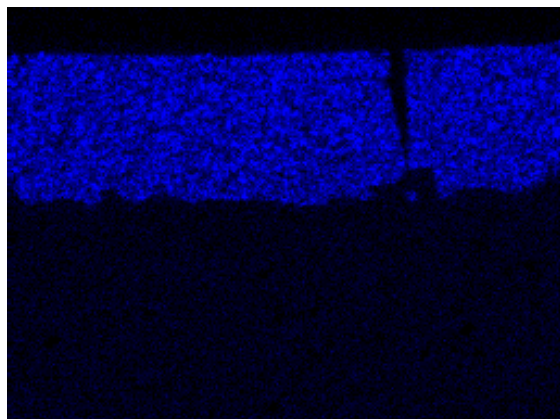


Figure 2. Nickel Map of Anode-Electrolyte Interface after 1,200-hr Stack Test

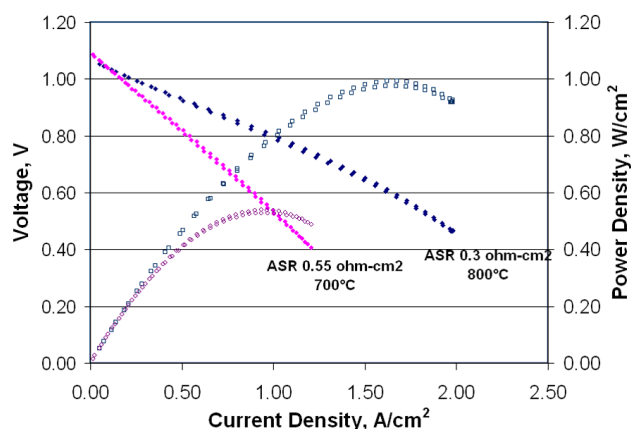


Figure 3. Performance of a Cathode-Supported LSGM Cell; Electrolyte Thickness of 75 Microns Was Used

microscopy of the anode-electrolyte interface showed no detectable diffusion of nickel into the electrolyte or an interfacial reaction product. The nickel x-ray map is shown in Figure 2.

Thin electrolyte single cells were fabricated using tape lamination technique. Both anode and cathode structures were evaluated as the support for the electrolyte. The performance of a cathode-supported cell is shown in Figure 3. The thin, 75-micron LSGM electrolyte cells showed an area-specific resistance (ASR) of 0.5 ohm-cm² at an operating temperature of 700°C. The long-term performance of selected cells is shown in Figure 4. Similar performance and stability results were also obtained using cells with the anode-support configurations. Thus, the performance benefits of using high-conductivity LSGM electrolyte and the

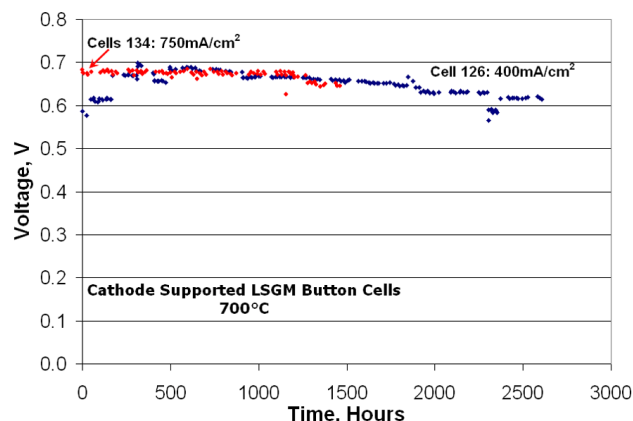


Figure 4. Long-Term Stability of Cathode-Supported Cells at an Operating Temperature of 700°C

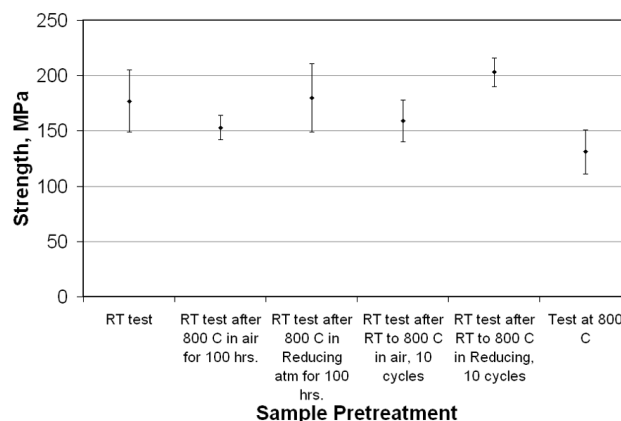


Figure 5. Strength Measurement of Gallate Bars at Room Temperature (RT)

stability improvement by using the modified anode were established in single-cell tests.

Mechanical characterization of gallate material was performed using ASTM bar samples. Both room-temperature and high-temperature strengths were evaluated. The results are summarized in Figure 5.

Conclusions

- Sr- and Mg-doped lanthanum gallate compositions show exceptionally high oxygen ion conductivity and stability in SOFC operating conditions for use at the intermediate temperatures of 650 – 700°C.
- The anode reactivity that is known to cause long-term instability has been addressed by introducing a modification to the nickel-based anode composition.

- A 1,000-hr stack test provided additional confirmation on the effectiveness of the anode modification.
- Thin, supported cells meet the performance target of 0.5 ohm-cm² resistance at 700°C.
- Long-term tests of single cells show stable performance.
- Mechanical tests show that the gallate material has adequate strength. Further improvement can be made by fabrication process improvements.

FY 2005 Presentations

1. Intermediate Temperature SOFC Operation Using Lanthanum Gallate Electrolyte, S. Elangovan, S. Balagopal, J. Hartvigsen, M. Timper, and D. Larsen, 29th International Conference on Advanced Ceramics and Composites, Second International Symposium on Solid Oxide Fuel Cells: Materials And Technology, January 2005.
2. Intermediate Temperature SOFC Operation Using Lanthanum Gallate Electrolyte, SECA Core Technology Program Review, Tampa, FL, January 27, 2005.